Supplementary Figure 1. Absorption spectrum of the SWNT network film deposited on a glass substrate. We see that the absorption peaks for the first and second subband transitions $E_{11}$ and $E_{22}$ are observed at 0.67 and 1.15 eV, respectively. The average diameter of this SWNT network film is estimated to be approximately 1.6 nm from the empirical Kataura plot\(^1\).
Supplementary Figure 2. Top view and cross-sectional SEM images of an MoO$_x$-SWNT/Si solar cell. The MoO$_x$ layer was thermally vapor deposited for 60 nm.
Supplementary Figure 3. Current density–voltage ($J$–$V$) curves for MoO$_x$-SWNT/Si solar cells as a function of the MoO$_x$ layer thickness.
Supplementary Figure 4. PCE, FF, current density, and open-circuit voltage as a function of the MoO$_x$ layer thickness.
Supplementary Figure 5. Experimentally obtained $J$–$V$ curves before (black circles) and after (red circles) the deposition of the MoO$_x$ coating on the SWNT film in the $p$-SWNT/$n$-Si solar cells. The $J$–$V$ curves calculated by Supplementary Equation (1) in Supplementary Note 1 are shown as solid lines.
Supplementary Figure 6. $J-V$ curves for $p$-SWNT/$n$-Si solar cells without (black line) and with insertion of an MoO$_x$ layer (red line) only between the SWNT film and the Au contact. Schematics of the device structures are shown in the figure. SWNT/Si solar cells with and without insertion of an MoO$_x$ layer between the SWNT network and the Au contact were fabricated to understand the effects of the MoO$_x$ as a hole transport layer. The pristine-SWNT/Si solar cell is found to have a $J_{sc}$ of 29.4 mA/cm$^2$, an FF of 65%, and a $V_{oc}$ of 0.58 V, which resulted in a $\eta$ of 11.2%. For the MoO$_x$-SWNT/Si solar cell, the $J_{sc}$, FF, and $V_{oc}$ were 31 mA/cm$^2$, 78%, and 0.59 V, respectively, and a high $\eta$ of 14.2% is obtained. The large enhancement in the PCE mainly results from an increase in the short-circuit current density and FF values, because the MoO$_x$ layer plays a crucial role in reducing the energy barrier for the charge carrier (hole) extraction and transport over the interface between the SWNT film and the Au contact, as discussed in the main text.
Supplementary Figure 7. $J–V$ curves for the $p$-SWNT/$n$-Si solar cells before (black line) and after (red line) the deposition of an MoO$x$ layer only on the active area. Schematics of the device structures are shown in the figures on the right. SWNT/Si solar cells with and without an MoO$x$ layer were fabricated only on the active area of the devices (red dashed square part in Supplementary Figure 7). The short-circuit current density and PCE of the pristine-SWNT/Si solar cell are $29 \text{ mA/cm}^2$ and $11.6\%$, respectively. For the MoO$x$-SWNT/Si solar cell; however, the short-circuit current density $J_{sc}$ drastically increases to $35 \text{ mA/cm}^2$, which results in a high value of $14.6\%$ for the PCE. The significant increase in the short-circuit current density and PCE is due to the deposition of the MoO$x$ on the active area of the cell as an antireflection layer for solar light and a carrier dopant for the SWNT network.
Supplementary Figure 8. IQE of the pristine-SWNT/Si (black line) and MoO$_x$-SWNT/Si (red line) solar cells.
Supplementary Figure 9. Raman spectra of SWNT network films without (black dot line) and with (red dot line) the MoO\textsubscript{x} coating deposited by evaporation. Inset shows the zoom of the Raman spectra from 1580 to 1620 cm\textsuperscript{-1}. The Raman spectra of SWNT network films were measured before and after MoO\textsubscript{x} layer coating. The Raman peak at \(~1600\) cm\textsuperscript{-1} corresponds to the G-band of SWNTs. The remarkable high frequency shift of the G-band (~3 cm\textsuperscript{-1}) is observed because of the presence of the MoO\textsubscript{x} layer coating. The high frequency shift of the G-band suggests the presence of the hole doping for SWNTs; this is consistent with the previously reported result\textsuperscript{4}. 
**Supplementary Figure 10.** The total resistivity ($R_{\text{tot}}$) as a function of distance $d$ for SWNT/Au (black circles) and MoO$_x$-SWNT/Au (red circles). The solid lines show the fitted results using Supplementary Equation (3) with a least-square fitting method. Inset shows the schematic of the TLM measurements.
**Supplementary Figure 11.** The schematic diagram of the equivalent circuit model of a device connected with a load, series resistance ($R_s$), shunt resistance ($R_{sh}$), and a diode ($D$).
Supplementary Figure 12. Carrier decays of SWNT/Si without and with the MoO\(_x\) coating. The minority carrier lifetimes of the SWNT on the \(n\)-Si substrate with and without the MoO\(_x\) layer were measured by the microwave-photo-conductivity decay method (\(\mu\)-PCD) to understand the effect of the Si substrate surface passivation. The minority carrier lifetimes with the MoO\(_x\) layer (MoO\(_x\)-SWNT/Si) and without the MoO\(_x\) layer (SWNT/Si) are estimated as 61 and 68 \(\mu\)s from the measured decay curves, respectively, and are almost unchanged by the MoO\(_x\) coating. This result indicates that the MoO\(_x\) layer does not strongly affect the surface passivation of the \(n\)-Si substrate.
Supplementary Figure 13. Photovoltaic efficiencies of 60 MoO₃-SWNT/Si solar cells. The photovoltaic power conversion efficiency is 16.8 ± 0.6%, which shows the very high reproducibility of this type of solar cell.
Supplementary Figure 14. (a) $J-V$ curves and (b) IPCE spectra of the different structures of the $n$-SWNT/$p$-Si heterojunction solar cells. The schematics of the device structures are shown in the figure, original (black line), ZnO deposited as antireflection layer (orange line), and as a multifunctional layer (red line).
Supplementary Figure 15. Top view and cross-sectional SEM images of $n$-SWNT/$p$-Si solar cells after the spin-coating of ZnO.
**Supplementary Table 1.** Parameters for the p-SWNT/n-Si solar cells without and with the MoO$_x$ coating. The analysis as presented in Supplementary Figure S5.

<table>
<thead>
<tr>
<th>Sample</th>
<th>$J_{sc}$ (mA/cm$^2$)</th>
<th>FF (%)</th>
<th>$V_{oc}$ (V)</th>
<th>$\eta$ (%)</th>
<th>$n$</th>
<th>$R_s$ (Ω)</th>
<th>$R_{sh}$ (KΩ)</th>
</tr>
</thead>
<tbody>
<tr>
<td>SWNT/Si</td>
<td>27.9</td>
<td>69</td>
<td>0.57</td>
<td>11.1%</td>
<td>1.24</td>
<td>360</td>
<td>8</td>
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<tr>
<td>MoO$_x$-SWNT/Si</td>
<td>36.6</td>
<td>78</td>
<td>0.59</td>
<td>17.0%</td>
<td>1.08</td>
<td>117</td>
<td>20</td>
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</table>
**Supplementary Table 2.** Variation of photovoltaic performance of SWNT/Si solar cell with MoO₃ coating.

<table>
<thead>
<tr>
<th>Samples</th>
<th>$J_{sc}$ (mA/cm²)</th>
<th>FF</th>
<th>$V_{oc}$ (V)</th>
<th>η (%)</th>
</tr>
</thead>
<tbody>
<tr>
<td>No. 1</td>
<td>36.6</td>
<td>0.78</td>
<td>0.59</td>
<td>17.0</td>
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<tr>
<td>No. 2</td>
<td>35.8</td>
<td>0.76</td>
<td>0.58</td>
<td>15.8</td>
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<tr>
<td>No. 3</td>
<td>34.5</td>
<td>0.80</td>
<td>0.59</td>
<td>16.3</td>
</tr>
<tr>
<td>No. 4</td>
<td>37.1</td>
<td>0.80</td>
<td>0.59</td>
<td>17.6</td>
</tr>
<tr>
<td>No. 5</td>
<td>36.9</td>
<td>0.79</td>
<td>0.59</td>
<td>17.2</td>
</tr>
</tbody>
</table>
Supplementary Table 3. The obtained values of $R_c$, $R_{\text{sheet}}$, $L_T$, and $\rho_c$ for SWNT/Au and MoO$_x$-SWNT/Au. The analysis and discussion as presented in Supplementary Figures S10 and Supplementary Note 4.

<table>
<thead>
<tr>
<th>Sample</th>
<th>$R_c$ (Ω)</th>
<th>$R_{\text{sheet}}$ (Ω/□)</th>
<th>$L_T$ (cm)</th>
<th>$\rho_c$ (Ω·cm$^2$)</th>
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<tbody>
<tr>
<td>SWNT/Au</td>
<td>155</td>
<td>3760</td>
<td>0.008</td>
<td>0.24</td>
</tr>
<tr>
<td>MoO$_x$-SWNT/Au</td>
<td>19</td>
<td>790</td>
<td>0.005</td>
<td>0.02</td>
</tr>
</tbody>
</table>
Supplementary Table 4. Parameters for the $n$-SWNT/$p$-Si solar cells without and with a ZnO coating.

$^a$ ZnO coating on the $n$-SWNT/$p$-Si solar cell as an antireflection layer.

$^b$ ZnO coating on the $n$-SWNT/$p$-Si solar cell as antireflection and electric transport layers.

<table>
<thead>
<tr>
<th>Sample</th>
<th>$J_{sc}$ (mA/cm$^2$)</th>
<th>FF (%)</th>
<th>$V_{oc}$ (V)</th>
<th>$\eta$ (%)</th>
</tr>
</thead>
<tbody>
<tr>
<td>$n$-SWNT/$p$-Si</td>
<td>23.6</td>
<td>20%</td>
<td>0.28</td>
<td>1.3</td>
</tr>
<tr>
<td>ZnO-SWNT/Si $^a$</td>
<td>26.2</td>
<td>22%</td>
<td>0.29</td>
<td>1.7</td>
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<tr>
<td>ZnO-SWNT/Si $^b$</td>
<td>31.9</td>
<td>29%</td>
<td>0.43</td>
<td>4.0</td>
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</table>
Supplementary Note 1: Analysis of the $J–V$ curves

The $J–V$ curves were analyzed using the equivalent circuit model to understand the photovoltaic properties of the prepared devices.

The total current density $J$ in this model can be described as follows.

$$J = J_0 \left\{ \exp \left( \frac{e(V - R_J)}{n k_B T} \right) - 1 \right\} + \frac{V - R_J}{R_{sh}} J_{ph},$$

(1)

where $J_0$ is the reverse saturation current density, $e$ is the electron charge, $n$ is the diode ideality factor, $k_B T$ is the thermal energy, $R_s$ is the series resistance, $R_{sh}$ is the shunt resistance, and $J_{ph}$ is the photocurrent density. Supplementary Figure 5 shows the experimental results for typical $J–V$ curves with (black circles) and without (red circles) the MoO$_x$ layer and the fitted $J–V$ curves (solid lines) obtained using Supplementary Equation (1). The fitted curves reproduce the experimental results well. The parameters, including the saturation current density ($J_{sc}$), FF, the open-circuit voltage, the PCE ($\eta$), the diode ideality factor ($n$), the series resistance ($R_s$), and the shunt resistance ($R_{sh}$), obtained from the $J–V$ curve analysis are shown in Supplementary Table 1.
Supplementary Note 2: Effect of MoO\textsubscript{x} as an antireflection layer

The IQE was then evaluated to understand the carrier doping effect of MoO\textsubscript{x} for the SWNT film. Figure S8 shows the IQE of the pristine-SWNT/Si (black solid line) and MoO\textsubscript{x}-SWNT/Si (red solid line) solar cells. The IQE for the MoO\textsubscript{x}-SWNT/Si solar cell is nearly identical to that of the pristine-SWNT/Si from 400 to 700 nm, but is greater above 700 nm. The increase in the short-circuit current density is calculated using the difference in the reflectivity of the pristine-SWNT/Si and the MoO\textsubscript{x}-SWNT/Si as follows:

\[ \Delta J = q \int \eta_c(E)(R_a - R_b)a(E)b_\lambda(E)dE, \]

where \( q \) is the electron charge, \( \eta_c(E) \) is the collected probability at a photon energy \( E \), \( R_a \) and \( R_b \) are the reflectivity of the SWNT/Si systems before and after MoO\textsubscript{x} coating, respectively, \( a(E) \) is the absorption, and \( b_\lambda(E) \) is the spectral photon flux density at \( E \). The increase in the current density calculated using Supplementary Equation (2) is 5.9 mA/cm\textsuperscript{2}, which is nearly the same as the experimentally obtained value of 6 mA/cm\textsuperscript{2} (Supplementary Figure 3).
Supplementary Note 3: Contact resistance of SWNT/Au and sheet resistance of SWNTs before and after MoO$_x$ coating

The contact resistance of SWNT/Au ($R_c$) and sheet resistance of SWNT ($R_{\text{sheet}}$) with and without the MoO$_x$ coating were measured by the transfer length method (TLM). We transferred the pristine SWNTs film and MoO$_x$-SWNTs film (with 2 mm width and 20 mm length) onto the glass substrate and evaporated Au contact pads using the mask technique. Inset of Supplementary Figure 10 shows the schematic of TLM measurements, where the distances ($d$) of the contact pads are 1, 2, 3, and 5 mm, and the length ($L$) and width ($W$) of contact pads are 2 and 2 mm, respectively. We measured the total resistance ($R_{\text{tot}}$) tested by the two-probe method between Au pads as a function of $d$ and potted the data in Fig. S11 for the SWNTs/Au (black circles) and MoO$_x$-SWNT/Au (red circles) samples. The total resistance is described as

$$R_{\text{tot}} = 2R_c + R_{\text{sheet}} \frac{d}{W}.$$  \hspace{1cm} (3)

The experimental data is accurately reproduced for SWNTs/Au and MoO$_x$-SWNT/Au, as shown in solid lines. We can evaluate the values of $R_c$ and $R_{\text{sh}}$ using the least-square fitting method. The contact resistance is equivalent to the resistance of an additional length of SWNTs sheet, and this equivalent length is called the current transfer length ($L_T$), and is given by

$$L_T = \frac{RW}{R_{\text{sheet}}}. \hspace{1cm} (4)$$

Moreover, the specific contact resistance $\rho_c$ is defined as

$$\rho_c = R_{\text{sheet}}L_T^2. \hspace{1cm} (5)$$

The obtained values of $R_c$, $R_{\text{sheet}}$, $L_T$ and $\rho_c$ are shown in Supplementary Table 3. The obtained current transfer length without the MoO$_x$ coating is almost the same as the previously reported value. The presence of the MoO$_x$ coating leads to drastic decreases of both $L_T$ and $\rho_c$ that are mainly caused by the decrease in the effective barrier height at the SWNT/Au interface due to the effect of MoO$_x$ as a hole transport layer and a dopant.

The reduction of the series resistance caused by the SWNT sheet resistance can be measured using...
\[ R_i = \frac{1}{8\pi} \rho_{\text{sheet}} / t, \]  

where \( R_i, t, \) and \( \rho_{\text{sheet}} \) are the series resistance, the SWNTs film thickness, and SWNTs sheet resistivity, respectively. We evaluate the series resistances from sheet resistance component as 150 ± 5 and 31 ± 3 Ω for the SWNT/Au and MoO\(_x\)-SWNT/Au, respectively.
Supplementary Note 4: Quantitative evaluation of PCE by MoO$_x$ coating in the devices.

We quantitatively evaluated the contribution of the MoO$_x$ coating to the PCE increase. The increase of PCE by the MoO$_x$ coating is estimated from the change of series resistance (1.), shunt resistance (2.) and diode loss (3.) from the equivalent circuit model, (Supplementary Figure 5) and reflection loss of solar light (4.).

1. The loss due to the series resistance can be described as,

$$ P_s = I_{\text{max}}^2 R_s, $$

where $P_s$ is the consumption power of series resistance, $I_{\text{max}}$ is the photocurrent at the maximum power point, and $R_s$ is the series resistance. The increase of PCE due to the change of the consumption power can be evaluated as,

$$ \Delta \eta = \frac{P_1 - P_2}{P_{\text{in}}} \times 100\%, $$

where $\Delta \eta$ is the PCE increase, $P_1$ and $P_2$ are the power consumptions before and after the MoO$_x$ coating, respectively, and $P_{\text{in}}$ is the input power. The series resistance including the components of SWNT film sheet resistance and the contact resistance at the SWNT/Au interface drastically decreases from 360 (2.83) to 117 $\Omega$ (0.92 $\Omega \cdot \text{cm}^2$), resulting in the total PCE improvement of 2.08% due to the sheet resistance and contact resistance reduction by the MoO$_x$ coating. To further understand the contributions of the sheet resistance and contact resistance components, the series resistances reduction by the sheet resistance and contact resistance contributions were calculated in supplementary 3, and were shown to result in the PCE increase of 0.95% and 1.13%, respectively.

2. The MoO$_x$ coating also increases the shunt resistance, which reduces the loss from the shunt current leakage. The loss contribution of the shunt resistance can be described as,

$$ P_{\text{sh}} = U_{\text{sh}}^2 / R_{\text{sh}}, $$

$$ U_{\text{sh}} = U_{\text{max}} + U_s, $$

$$ U_s = I_{\text{max}} R_s, $$
where $P_{sh}$, $U_{sh}$, and $I_{sh}$ are the shunt resistance consumption power, shunt voltage, and shunt current, respectively, $U_{max}$ and $I_{max}$ are the load voltage and current, respectively, and $U_s$ is the series voltage. Using Supplementary Equation (8) – (11), the PCE increase due to the shunt resistance changes induced by the presence of the MoO$_x$ coating was determined to be 1.46%.

3. The power of the diode is calculated as

$$P_D = U_D I_D,$$

$$U_D = U_{max} + U_s,$$

$$I_D = I_{sc} - I_{sh} - I_s,$$

where $P_D$, $U_D$, and $I_D$ are the consumption power of power, diode voltage, and current flow through diode, respectively. $U_D$ and $I_{sc}$ are the load voltage and open circuit current, respectively. The PCE increase due to the reduction of the diode loss was evaluated as about 0.09%.

4. We estimated that the reduction of the reflected solar light by the antireflection effect increases the current density by 5.9 mA/cm$^2$ as described above (Supplementary Note 3 and Supplementary Figure 7). This results in the PCE increase of 2.25%.
Supplementary Note 5: Fabrication of $n$-SWNT/$p$-Si solar cells

For the $n$-SWNT/$p$-Si solar cells, $p$-Si substrates (2–5 Ω-cm) were used. The fabrication procedure is nearly the same as that for the $p$-SWNT/$n$-Si cell described in the main text. An aqueous solution of β-nicotinamide adenine dinucleotide (NADH, 90 mM, 10 μl) was deposited on the SWNT network film for $n$-type doping in the glove box. An aqueous colloidal ZnO solution was prepared as previously reported and spin-coated on the SWNT film. The detailed device structure for the $n$-SWNT/$p$-Si solar cell is shown in the middle of Supplementary Figure 14.

The morphology of a ZnO-coated NADH (nicotinamide adenine dinucleotide with hydrogen) doped SWNT film is shown in Supplementary Figure 15. Nanorods with a size of several tens of nanometers can be observed to uniformly cover the substrate. From the cross-sectional SEM image, the thickness of the ZnO layer and the NADH film are estimated to be approximately 70 and 20 nm, respectively.
Supplementary References


